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Short communication

High performance solid oxide electrolysis cell with impregnated electrodes

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ABSTRACT

Here we report a solid oxide electrolysis cell (SOEC) employing impregnated electrodes. The cell structure consisted of a porous 430 L metal support, a Ni-$Ce_{0.8}Sm_{0.2}O_{2-\delta}$ (SDC) impregnated 430 L-zirconia stabilized zirconia (YSZ) hydrogen electrode, a scandia stabilized zirconia (SSZ) electrolyte and a Nd_2O_3-$Nd_2NiO_{4+\delta}$ (Nd_2O_3-NNO) impregnated SSZ oxygen electrode. The cell is prepared by tape casting, co-firing and impregnation techniques. At an applied voltage of 1.3 V and 50% steam content, current density of 2.05 A cm^{-2} is obtained at 800 °C. The effect of the variation of H_2O/H_2 ratio (3/97 to 70/30) on electrolysis performance at 750 °C is evaluated and the long-term stability in electrolysis mode is also investigated.

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1. Introduction

Hydrogen is a clean energy carrier with the highest weight specific energy density and has been considered as potentially the best future alternative to fossil fuels. Of all known methods to generate hydrogen, water electrolysis is probably the cleanest method and produces hydrogen with high purity and no pollution [1,2]. Water electrolysis involves both the hydrogen evolution reaction at the cathode and the oxygen evolution reaction at the anode. The theoretical water splitting voltage is 1.23 V at 25 °C and 0.95 V at 900 °C [1]. Operating at high temperature decreases the water splitting voltage and the electrical energy demand due to lower polarization and facile reaction kinetics. Both thermodynamic and kinetic factors result in higher current density at a given cell voltage and steam partial pressure [3]. High temperature steam electrolysis to hydrogen using solid oxide electrolysis cells (SOECs), which is defined as the reverse of solid oxide fuel cells (SOFCs), has shown higher energy efficiency for hydrogen production when compared to alkaline or proton exchange membrane (PEM) water electrolysis [4].

Research into SOECs has increased significantly in recent years. Tietz et al. reported a hydrogen electrode supported SOEC with a current density of −1 A cm^{-2} at an absolute humidity of 80 vol.% which displayed an overall degradation rate of only 3.8% kh^{-1} during operation at about 778 °C for 9000 h [5]. Ebbesen et al. also studied a Ni/YSZ electrode supported SOEC stack operating at −0.25 A cm^{-2} at 850 °C in a 1:1 atmosphere of H_2O/H_2 for 830 h, demanding that long-term electrolysis is feasible without notable degradation [6]. Despite these promising results, the development of SOECs is still at the lab-scale and stability issues such as delamination at the oxygen electrode/electrolyte interface, coarsening and agglomeration of nickel particles require further refinement [7]. Preparing cell electrodes *via* the impregnation method can help overcome these issues [8–10]. Although the low mechanical strength of ceramic backbones with high porosity could restrict the development of such techniques, metal-supported solid oxide fuel cells (MS-SOFCs) using a mechanically robust metal to support the whole cell could effectively solve this problem [11].

In this work, we developed an intermediate temperature (650 °C–800 °C) SOEC comprising impregnated electrodes and a metal support. The cell consisting of a porous 430 L metal support, a Ni-SDC impregnated 430 L-YSZ hydrogen electrode, SSZ electrolyte and a Nd_2O_3-NNO impregnated SSZ oxygen electrode, was prepared by tape casting, co-firing and impregnation techniques. The electrochemical performance at varying temperatures and steam contents were investigated and the long-term stability in SOEC mode was also studied.

2. Experimental

The metal-supported solid oxide electrolysis cell (MS-SOEC) was fabricated *via* a two-step procedure. Firstly, the four-layer structure—porous 430 L (strainless steel) support | porous 430 L-$8Zr_{0.92}Y_{0.08}O_{1.96}$ (YSZ) backbone | $Zr_{0.88}Sc_{0.22}Ce_{0.01}O_{2.12}$ (SSZ) electrolyte | porous SSZ backbone—was prepared by laminating four tape-casted green tapes, followed by co-firing in a reducing atmosphere (5% H_2–95% N_2) at 1300 °C for 4 h [12]. Secondly, the oxygen electrode catalyst, Nd_2O_3-$Nd_2NiO_{4+\delta}$ (Nd_2O_3-NNO), was introduced into the porous SSZ backbone using an aqueous solution containing $Nd(NO_3)_3$, $Ni(NO_3)_2$ and citric acid in a molar ratio of 2:1:3. After drying, the Ni-$Sm_{0.2}C_{0.8}O_{2-\delta}$ (SDC) was impregnated into the porous 430 L-YSZ backbone as

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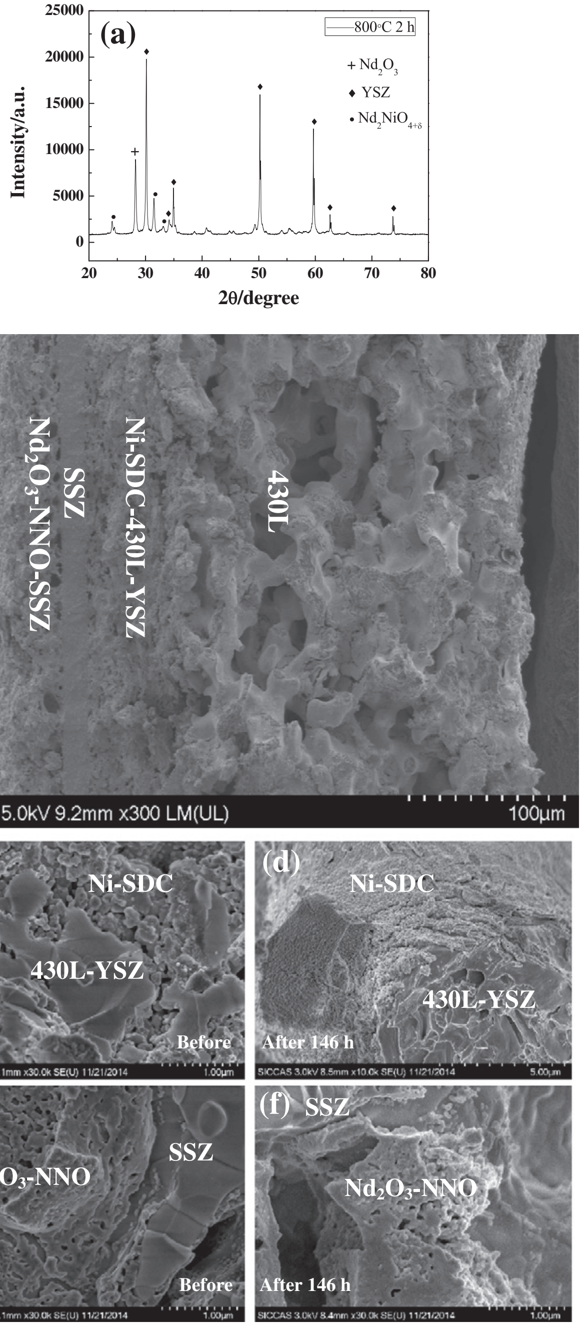

Fig. 1. (a): XRD pattern of the $Nd_2NiO_{4+\delta}$ impregnated YSZ composite measured at 800 °C. (b): SEM micrograph of the cross-section of the MS-SOEC. (c) and (d): Ni-SDC impregnated 430 L-YSZ composite hydrogen electrode before and after long-term durability electrolysis tests. (e) and (f): NNO impregnated SSZ composite oxygen electrode before and after long-term durability electrolysis tests.

the hydrogen electrode catalyst at a weight ratio of 2:8 [13]. Both catalysts were impregnated into the porous SSZ backbone and porous 430 L-8YSZ backbone for each experiment. Heat treatment of the electrodes was conducted at 350 °C in air for 0.5 h. The loadings of impregnated catalysts were controlled using a micro-liter syringe and the impregnation/heat treating cycle was repeated to achieve the desired loadings (30 wt.% catalyst loading for the oxygen electrode and 10 wt.% catalyst loading for the hydrogen electrode). The final sintering of the impregnated catalysts was conducted at 800 °C for 2 h in-situ before the cell testing process. The desired NNO phase was confirmed by X-ray diffraction as shown in Fig. 1(a). Although the Nd_2O_3 phase is observed, previous studies have shown that it does not affect the cathode performance [14,15].

For electrochemical measurements, silver paste was painted onto the electrode surfaces as the current collector. Steam was produced using a bubbler with precise temperature control and the ratio of steam to the fuel gas (H_2) was controlled by setting the bubbler temperature according to saturated steam pressure curve. The effective oxygen electrode active area of the single cell was 0.35 cm^2. The cell was evaluated in SOEC mode with different partial pressures of H_2O/H_2 (3/97-70/30) and applied voltages (0.3–1.6 V) at a scan rate of 20 mV s^{-1}. Electrochemical impedance spectra (EIS) were measured with an IM6 Electrochemical Workstation (ZAHNER, Germany) employing a sinusoidal amplitude signal of 20 mV in the frequency range of 50 mHz to 100 kHz. Microstructures of the SOEC were examined by scanning electron microscopy (SEM) using a Hitachi SU-8220 microscope.

3. Results and discussion

Fig. 1(b) shows the cross-sectional SEM micrograph of SOEC containing a porous Nd_2O_3-NNO impregnated SSZ oxygen electrode (≈30 µm), a dense SSZ electrolyte (≈15 µm), a porous Ni-SDC impregnated YSZ-430 L composite hydrogen electrode (≈50 µm) and a porous 430 L substrate (≈200 µm). Good adherence between the different

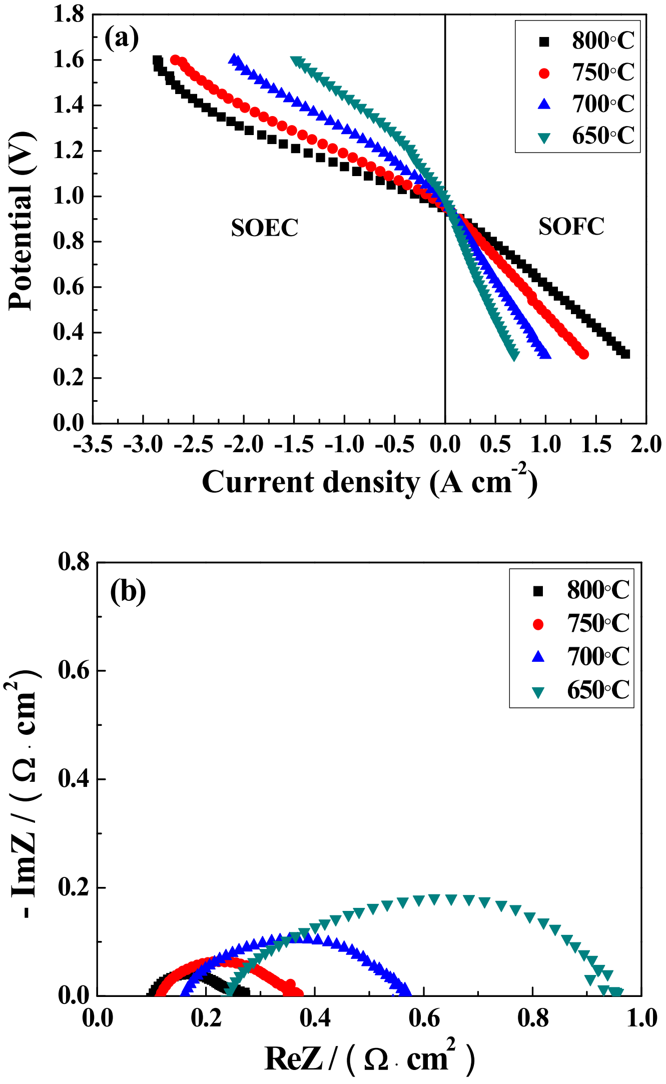

Fig. 2. (a): I–V curves of the single cell in electrolysis mode (H_2O/H_2 = 50/50) measured at 650 °C–800 °C. (b): Electrochemical impedance spectra measured at open circuit potential.

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